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TRANSPORT PROPERTIES OF PARTIALLY IONIZED NITROGEN

I. THE COLLISION INTEGRALS

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THEORETICAL FLUID PHYSICS SECTION

TRANSPORT PROPERTIES OF PARTIALLY IONIZED NITROGEN

I. THE COLLISION INTEGRALS

By

M. P. Sherman

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ABSTRACT

This report is the first of two concerned with the computation of the transport properties of nitrogen up to temperatures where there is single ionization. In this report the collision integrals are obtained using the best available information. In particular, use is made of the recent work of Knof, Mason, and Vanderslice for the charge exchange cross section of the $N - N^+$ interaction. Calculations were performed to obtain the $\Omega^{(2,2)}$ collision integral for the $N - N^+$ interaction.

In a second report, transport properties will be computed by a new method which permits rapid calculation of the transport properties of multi-component mixtures valid up to the second approximation of the variational scheme of Hirschfelder, et. al.

Author

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SYMBOLS

A	=	Nondimensional energy used in e - N ₂ analysis
A^*	=	Nondimensional ratio of collision integrals $\frac{\Omega^{(2,2)}}{\Omega^{(1,1)}}$
a	=	Parameter used in exponential potential
B	=	Nondimensional energy used in e - N ₂ analysis
B^*	=	Nondimensional ratio of collision integrals
b	=	Impact parameter
C^*	=	Nondimensional ratio of collision integrals
c	=	Speed of light
D_e	=	Dissociation energy
e	=	Electrons
e	=	Electronic charge 4.80×10^{-10} E.S.U.
E	=	Initial kinetic energy of collision
$I^{(22)}$	=	Tabulated function used with the exponential potential
h	=	Planck constant
h	=	Debye length
k	=	Boltzmann constant
\ln	=	Natural logarithm
\log	=	Logarithm to base 10
m_i	=	Particle mass
M_i	=	Particle molecular weight
n	=	Total particle density
n_i	=	Particle density of species i
N_2	=	Nitrogen molecule
N	=	Nitrogen atom
N^*	=	Electronically excited nitrogen atom
N^+	=	Singly ionized nitrogen atom

$Q^{(\ell)}$	=	Collision cross section of type ℓ , $\ell = 0, 1, 2 \dots$
\tilde{Q}, \tilde{Q}'	=	Integral quantities defined in Ref. 1
r	=	Interparticle distance
r_e	=	Interparticle distance of potential minimum
r_m	=	Minimum interparticle distance in collision
T	=	Temperature
$T^* = kT/\epsilon$	=	Nondimensional Temperature
$Z^{(2,2)}$	=	Tabulated function used with Buckingham potential
Z_i	=	Unit electronic charge of particle of species i (i.e. $Z_{N^+} = 1$, $Z_e = -1$)
α	=	Polarizability
α	=	Parameter in Buckingham (Exp. 6) potential
α	=	Parameter used with exponential potential
Γ_a^b	=	$\int_a^b e^{-x^2} dx$
δ	=	$\sqrt{\frac{1}{2} \mu_g^2 / kT}$, δ^2 is a nondimensional initial kinetic energy of collision
Δ	=	e^2 / kT
ϵ	=	Depth of the potential minimum of an interparticle potential curve
μ	=	Reduced mass
${}^6\Pi_u$	=	Pi molecular state of multiplicity 6 (x2), Ungerade-Standard spectroscopic notation
ρ	=	Parameter used in exponential potential
${}^2\Sigma_g^+$	=	Sigma plus state of multiplicity 2, Gerade-Standard spectroscopic notation
ϕ	=	Interparticle potential

χ = Angle of deflection of particles in the center of gravity
coordinate system

$\Omega^{(\ell, s)}$ = Collision integral (Chapman Enskog form)

$\Omega^{*(\ell, s)}$ = Nondimensional collision integral (Hirschfelder, et. al.)

$\overline{\Omega^{(\ell, s)}}$ = Collision integral form used in this work (in A^2)

1. INTRODUCTION

This report is the first of two considering the computation of the transport properties of partially ionized nitrogen, i.e., mixtures of nitrogen molecules, (N_2), nitrogen atoms, (N) electronically excited atoms, (N^*), ions, (N^+), and electrons, (e). In this report the collisional information used in the theory is presented, while in the second, the transport property calculations will be presented. The calculations can be considered applicable up to conditions where the effects of double ionization begin to be significant.

The nitrogen system was chosen because nitrogen is the principal constituent of the earth's atmosphere and probably that of Mars and Venus. Further, there has been some controversy on hypersonic heat transfer in nitrogen, which hinges on disagreements on the transport properties of partially ionized nitrogen, [1]*.

It is hoped that this work will be more accurate than previous works because of the use of the best available knowledge of the collisional information and the use of a new formalism which rapidly computes transport properties using the Chapman-Enskog method [2] valid up to the second approximation of the variational scheme of Hirschfelder [3]. In particular, use is made of the new work on the $N - N^+$ interaction of Knof., Mason, and Vanderslice [4] for the diffusion collision integral $\Omega^{(1,1)}$, and computations of the author for the $\Omega^{(2,2)}$ collision integral.

The collisional information for the Chapman-Enskog theory appears in the form of "collision integrals." The collision integrals involve certain weighted collision cross sections averaged over the zeroth (Maxwellian) approximation of the velocity distribution function of the colliding partners. The notation used in the main follows Hirschfelder, et.al. [3] rather than Chapman and Cowling [2].

*Numbers in brackets indicate references.

$$\chi(g, b) = \pi - 2b \int_{r_m}^{\infty} \frac{dr}{r^2 \sqrt{1 - \frac{b^2}{r^2} - \frac{\varphi(r)}{\frac{1}{2} \mu g^2}}} \quad (1)$$

$$Q^{(\ell)} = 2\pi \int_0^{\infty} (1 - \cos^{\ell} \chi) b db \quad (2)$$

$$\Omega^{(\ell, s)} = \sqrt{\frac{kT}{2\pi\mu}} \int_0^{\infty} \exp(-\delta^2) \delta^{2s+3} Q^{(\ell)} d\delta \quad (3)$$

$$\overline{\Omega^{(\ell, s)}} \equiv \sigma^2 \Omega^{*(\ell, s)} = \frac{\sqrt{2\pi\mu/kT} \Omega^{(\ell, s)}}{\frac{1}{2} (s+1)! \left[1 - \frac{1}{2} \frac{1 + (-1)^{\ell}}{1 + \ell} \right] \pi} \quad (4)$$

$$A^* = \overline{\Omega^{(2, 2)}} / \overline{\Omega^{(1, 1)}} \quad (5)$$

$$B^* = \left[\overline{5 \Omega^{(1, 2)}} - \overline{4 \Omega^{(1, 3)}} \right] / \overline{\Omega^{(1, 1)}} \quad (6)$$

$$C^* = \overline{\Omega^{(1, 2)}} / \overline{\Omega^{(1, 1)}} \quad (7)$$

$$\delta^2 \equiv \frac{\frac{1}{2} \mu g^2}{kT} \quad (8)$$

$$\mu \equiv \frac{m_1 m_2}{m_1 + m_2} \quad (9)$$

The quantity $\overline{\Omega}$ rather than Ω^* will be used since the "equivalent rigid sphere diameter", σ , has no real meaning for many of the potentials considered. The integrals are convergent for all but the Coulombic potential. The divergence of the collision integral for the Coulombic potential has cast doubt on the validity of the Chapman-Enskog method for ionized gases.

However, it has been found that the method, when properly carried out, gives results equivalent to the Fokker-Planck equation used for plasmas. It therefore seems that the Chapman-Enskog method can be used for partially ionized gas mixtures.

The theory has been extended to include the effects of inelastic collisions [5], but thus far, there has been very little use of the extension to obtain numerical results. What is usually done is to use an Eucken-type approximation. Here, it is assumed that there is easy transfer of energy between internal and translational states. Further, it is usually assumed that the collision cross section is not affected by the internal energy state. The first assumption is valid essentially if there are few long-lived metastable states. The latter assumption is reasonably good for rotation and vibration, but usually not for electronic excitation.

The mole fraction of the excited electronic states can be significant. The electronic levels which are low enough so that their populations could be important are listed in Table 1.

The first two excited levels of N^+ are essentially ground states, since at the temperatures at which N^+ exists in quantity their excitation energy is small compared to the mean translational energy. The 1D excited state of N^+ is not important. The important electronically excited states are the 2D 2P levels of N . It can easily be shown that their concentration can be significant, e.g. at 11,000 K 19% of the nitrogen atoms are electronically excited. The still higher levels do not become important until higher temperatures, where the concentration of N rapidly decreases.

It has been suggested by Yos [7] that the elastic collision cross section of the 2D excited level is close to that of the ground state, since no electron has been excited to a higher principal quantum number. A discussion of the elastic collision cross sections of electronically excited atoms can be found in [8, 9]. However, there is an excitation exchange effect which could greatly increase $\Omega^{(1,1)}$, but not $\Omega^{(2,2)}$ [10]. Yos assumed this excitation exchange cross section was similar to the large charge exchange

cross section of the $N - N^+$ interaction. These assumptions are reasonable.

See Section 9.

The interactions which must be considered are shown in Table II.

2. THE NEUTRAL INTERACTIONS

For the neutral interactions, $N_2 - N_2$, $N - N$, and $N - N_2$, use has been made of the work of Mason and co-workers [11,12]. This information has been tabulated into a form directly applicable to transport property calculations [12].

The interaction of ground state nitrogen atoms can take place along any one of four potential curves. It has been shown that in the case of multiple interaction curves the collision integral is a weighted average of the collision integrals for each of the potential curves, the statistical weight being the relative probability of collisions of the type [10]. The result of the analysis shows that the main contribution to the collision integral does not come from the fairly well known ground state curve of the molecule but rather from the higher nonbinding potential curves.

The potentials of the other two interactions are not spherically symmetric, but they are averaged over the angular orientation. It should be noted that extrapolation of low temperature data on the $N_2 - N_2$ interaction can lead to error since portions of the potential curve of little importance at low temperatures, and therefore usually only poorly known, are important at high temperatures.

3. THE COULOMBIC INTERACTIONS

The conceptual difficulties with the handling of the Coulombic potential has been previously mentioned. The best model for handling these interactions appears to be the use of the shielded Coulomb potential

$$\phi = - \frac{Z_1 Z_2 e^2}{r} \exp (-r/h) \quad (10)$$

as done by Lieboff [13]. His results are shown below.

$$\overline{\Omega^{(1,1)}} = \frac{1}{2} \Delta^2 \left\{ \ln \left(\frac{2h}{\Delta} \right) - .961 \right\} \quad (11)$$

$$\overline{\Omega^{(2,2)}} = \frac{1}{2} \Delta^2 \left\{ \ln \left(\frac{2h}{\Delta} \right) - .461 \right\} \quad (12)$$

$$\overline{\Omega^{(1,2)}} = \frac{1}{6} \Delta^2 \left\{ \ln \left(\frac{2h}{\Delta} \right) + .039 \right\} \quad (13)$$

$$\overline{\Omega^{(1,3)}} = \frac{1}{12} \Delta^2 \left\{ \ln \left(\frac{2h}{\Delta} \right) + 1.039 \right\} \quad (14)$$

$$h = \sqrt{\frac{kT}{4\pi e^2 n_e}} = \text{Debye Length} \quad (15)$$

$$\Delta = \frac{e^2}{kT} \quad (16)$$

4. THE e - N₂ INTERACTION

Information on the collision cross section for the e - N₂ interaction has been obtained from electron scattering experiments at higher energies and mobility studies at lower energies. Collision cross section information was summarized in a recent article [14]. There is no information on Q⁽²⁾. It seems most reasonable to assume isotropic scattering so that

$$Q^{(0)} = Q^{(1)} = \frac{3}{2} Q^{(2)} \quad (17)$$

The experimental curve was closely fitted up to about 1 e.v., after which the fit is not close. This should be sufficient since one does not expect to find co-existing sizable amounts of molecular nitrogen and energetic electrons.

$$Q^{(1)} = \begin{cases} 10 & E > .274 \text{ e.v.} \\ 19 \sqrt{E} & .007 < E < .274 \text{ e.v.} \\ 1.6 & E < .007 \text{ e.v.} \end{cases} \quad (18)$$

$$\overline{\Omega^{(1,1)}} = \frac{1}{2\pi} \left\{ \begin{aligned} &1.6 [2 - \exp(-A)(A^2 + 2A + 2)] \\ &+ 10 \exp(-B)(B^2 + 2B + 2) \\ &+ 19 \sqrt{kT} \exp(-A) \left[A^2 + \frac{5}{2} A + \frac{15}{4} \right] \\ &- 19 \sqrt{kT} \exp(-B) \left[B^2 + \frac{5}{2} B + \frac{15}{4} \right] \\ &+ \frac{15}{4} \times 19 \sqrt{kT} \frac{\Gamma \sqrt{B}}{\sqrt{A}} \end{aligned} \right\} \quad (19)$$

$$\overline{\Omega^{(2,2)}} = \frac{2}{3\pi} \left\{ \begin{aligned} &1.6 [6 - \exp(-A)(A^3 + 3A^2 + 6A + 6)] \\ &+ 10 \exp(-B)(B^3 + 3B^2 + 6B + 6) \\ &+ 19 \sqrt{kT} \sqrt{A} \exp(-A) \left[A^3 + \frac{7}{2} \left(A^2 + \frac{5}{2} A + \frac{15}{4} \right) \right] \\ &- 19 \sqrt{kT} \sqrt{B} \exp(-B) \left[B^3 + \frac{7}{2} \left(B^2 + \frac{5}{2} B + \frac{15}{4} \right) \right] \\ &+ \frac{7 \times 15}{8} \times 19 \sqrt{kT} \frac{\Gamma \sqrt{B}}{\sqrt{A}} \end{aligned} \right\} \quad (20)$$

$$\overline{\Omega^{(1,2)}} = \overline{\Omega^{(2,2)}} \quad (21)$$

$$\overline{\Omega^{(1,3)}} = \frac{1}{24\pi} \left\{ \begin{aligned} &1.6 [24 - \exp(-A) (A^4 + 4A^3 + 12A^2 + 24A + 24)] \\ &+ 10 \exp(-B) [B^4 + 4B^3 + 12B^2 + 24B + 24] \\ &+ 19 \sqrt{kT} \sqrt{A} \exp(-A) \left[A^4 + \frac{9}{2} \left\{ A^3 + \frac{7}{2} \left(A^2 + \frac{5}{2} A + \frac{15}{4} \right) \right\} \right] \\ &- 19 \sqrt{kT} \sqrt{B} \exp(-B) \left[B^4 + \frac{9}{2} \left\{ B^3 + \frac{7}{2} \left(B^2 + \frac{5}{2} B + \frac{15}{4} \right) \right\} \right] \\ &+ \frac{7 \times 9 \times 15}{16} \times 19 \sqrt{kT} \frac{\sqrt{B}}{\sqrt{A}} \end{aligned} \right\} \quad (22)$$

where

$$A = \frac{.007}{kT}, \quad B = \frac{.274}{kT}, \quad k = 8.514 \times 10^{-5} \text{ e.v./}^\circ\text{K} \quad (23)$$

$$\Gamma_A^B \equiv \int_A^B \exp(-x^2) dx$$

5. THE e - N INTERACTION

The experimental measurement of the e - N collision integral is a much more difficult task than that of the e - N₂ collision integral. However, the problem is more amenable to theoretical analysis, although still difficult. There is considerable disagreement between the various published results [15, 16, 17]. It was decided to use the most recent work, that of Klein and Brueckner [15]. Their result shows a cross section which does not vary much at the energies of interest. In view of the uncertainties, the cross section was approximated by a constant. The scattering is assumed isotropic a good assumption for low energy electrons scattered by atoms.

$$\overline{\Omega^{(1,1)}} = \frac{13}{\pi} \quad (24)$$

$$A^* = B^* = C^* = 1 \quad (25)$$

6. THE N - N⁺ INTERACTION

The N - N⁺ interaction will have a very important effect on the value of the transport properties when the nitrogen is partly ionized. Furthermore, the value for the collision integrals for this interaction has been quite uncertain. Therefore, it will be discussed in detail. The value of $\Omega^{(1,1)}$ for this inter-action was computed by Knof, Mason, and Vanderslice [4]; the value of $\Omega^{(2,2)}$ was computed by the author based on the potential curves of [4, 25].

In the interaction of an atomic ion and its parent atom there is a symmetric charge exchange effect. The process involves no interchange of translation and internal energy and can be treated as an elastic collision except that the angle of deflection which would normally be χ appears to be $\pi - \chi$. See Figure 1. Hence, for collisions with large impact parameters the apparent effect of charge exchange is to change the normally expected small deflection into nearly a 180 degree deflection. Although the apparent deflection has been altered by the change in the identity of the particles, the momentum transfer caused by the charge (or excitation) exchange is negligible. Hence, one might intuitively expect that the cross section associated with diffusion, $Q^{(1)}$, would be altered, but that the cross section associated with viscosity and frozen translational thermal conductivity, $Q^{(2)}$, would not. This has been shown to be true [10].

The charge on the ion will induce a dipole moment on the atom. The resultant potential is of the form

$$\phi(r) = - \frac{\alpha e^2}{2r^4} \quad (26)$$

$$\alpha = \text{polarizability (1.13 A}^3 \text{ for N)}$$

The inverse fourth power potential, being the slowest decreasing potential in this interaction, must dominate at large interparticle distances. Hence, its effect will dominate in the low temperature limit.

Some authors have assumed the charge induced dipole effect dominates at all temperatures, while others have assumed it does not affect the collision integral at any but the lowest temperatures. The latter is correct.

Given the potential curves, or rather the separation of the g (Gerade) and u (Ungerade) states, the charge exchange cross section can be calculated. The method is discussed in [4]. Their result is [4].

$$\pi \overline{\Omega^{(1,1)}} = 102.4 - 13.11 \frac{\text{Log}(T)}{10} + .415 \left[\frac{\text{Log}(T)}{10} \right]^2 \quad (27)$$

Note that the definition of the collision integral in [4] and the present work differ by a factor of π . The pure charge induced dipole collision integral is easily found using standard references for inverse power attractive forces [18, 19].

$$\pi \overline{\Omega^{(1,1)}} = \frac{1420}{T^{1/2}} \quad (28)$$

There must be some intermediate region where the effects of the charge induced dipole and charge exchange are both important. This transition region was not studied, but simple physical reasoning led to the conclusion that the collision integral in the intermediate region would be adequately represented by fairing the two asymptotic results with a smooth curve. The final result presented indicates that there is no effect of the charge induced dipole above about 1200 K.

For the $\overline{\Omega^{(1,1)}}$ calculation all that is necessary to obtain reasonably accurate charge exchange cross sections is the behavior of the potentials at far separations. The formulas for the potentials of [4] are quoted as being valid only beyond 1.95 Å. However for the calculation, since there is no charge exchange effect, one must know the potential curves in to the point where their potential energy is above the thermal kinetic energies of the particles. Fortunately a recent report by Gilmore [25] critically summarizes all the available information on the potential curves. Using [25] and [4] the collision integral was computed in the Appendix

Unfortunately neither of these two references considers the long range charge induced dipole effect. At the lowest temperatures we must have

$$\overline{\Omega^{(2,2)}} = \frac{393}{T \frac{1}{2}} \quad (29)$$

The difficulty is in the transition. This is discussed in more detail in the Appendix , but in the end the transition is not calculated but merely estimated.

7. THE $N_2 - N^+$ INTERACTION

At long range the dominant potential will be due to the charge induced dipole. The dielectric constant of nitrogen at S.T.P. is 1.000590 and therefore its polarizability is 1.73 \AA^3 . Hence, in the low temperature limit,

$$\overline{\Omega^{(1,1)}} = \frac{558}{T^{1/2}} \quad (30)$$

$$\overline{\Omega^{(2,2)}} = \frac{486}{T^{1/2}} \quad (31)$$

$$\overline{\Omega^{(1,2)}} = \frac{465}{T^{1/2}} \quad (32)$$

$$\overline{\Omega^{(1,3)}} = \frac{407}{T^{1/2}} \quad (33)$$

The potential for this interaction has been approximated by a 12-6-4, Eq. 30, potential by Woolley [21].

$$\phi = \frac{\epsilon}{2} \left[(1 + \gamma) \left(\frac{r_e}{r} \right)^{12} - 4\gamma \left(\frac{r_e}{r} \right)^6 - 3(1 - \gamma) \left(\frac{r_e}{r} \right)^4 \right] \quad (34)$$

ϵ = depth of potential minimum, .127 E. V.

r_e = internuclear distance of minimum, 3.02 A

γ = parameter, .22

Collision integrals for this potential are tabulated in [18].

8. THE EXCITED NITROGEN ATOM INTERACTIONS

There are twelve intermolecular potential curves for the interaction between a ground state nitrogen atom and the lowest electronically excited state of nitrogen, the 2D level. What information exists is best shown by Gilmore [25]. The curves will correspond to Σ^+ , π , and Δ states, triplets and quintets, with subscripts g and u [24]. The curves possessing potential wells are imperfectly known while the shapes of repulsive potentials have not been measured or calculated. Information for the next higher level, the 2P level, with eight intermolecular potentials, is even more scanty. Much the same condition exists for interactions between atoms in the 2D state. It is clear that more information will be necessary before elastic scattering and excitation exchange cross sections can be calculated.

The assumption of Yos [7] that the elastic scattering cross sections are not significantly affected by the electronic excitation seems reasonable, but until the curves are known and calculations made, or experiments conducted, the error introduced by the assumption is not known. There are no known calculations of the excitation exchange cross section between excited and ground state nitrogen atoms. It was the unanimous opinion of several consultants (see acknowledgements) that the total cross section involving excitation exchange lies between the normal gas kinetic and the larger charge exchange cross section, probably near the latter.

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APPENDIX

CALCULATION OF THE $\overline{\Omega^{(2,2)}}$ COLLISION INTEGRAL OF $N - N^+$

Computations Ignoring the Charge Induced Dipole Effect

Perhaps the best overall information on the intermolecular potentials of $N - N^+$ are the curves of Gilmore [25]. For separations of greater than about 1.95 Å there are the formulas of Knoff, Mason, and Vanderslice [4]. The formulas are reproduced in Table IV, where

$$2\tilde{Q} = 261.4 \exp(-2.977r) \quad r > 1.95 \text{ Å} \quad (A1)$$

$$2\tilde{Q}' = 675.8 \exp(-3.468r) \quad r > 1.95 \text{ Å} \quad (A2)$$

are given in [4] and we easily obtain

$$J\pi = \{185 \exp(-2.46r) + 196 \exp(-2.977r)\} \quad (A3)$$

$$J'\pi + J'\sigma = 1013.7 \exp(-3.468r) - 858 \exp(-2.46r) \quad (A4)$$

Multiplicity Six Repulsive Potentials

The multiplicity six potential curves have a total statistical weight half the total of all the potential curves. Since their individual cross sections are larger than those of the other potential curves, the contribution of the multiplicity six potentials will be much larger than half of the total. As indicated by Gilmore [25] the value of these potential curves is somewhat uncertain. They rise rapidly so that at 1.95 Å they are over three electron volts. Hence the formulas [4] should be sufficient for energies below this magnitude. The repulsive potentials then consist of the sum

of two exponential terms which can be remarkably well approximated by one exponential term. The approximation was done in a conventional manner by fitting a straight line through points plotted in semi-log paper. The potentials, valid for $r > 1.95$ Å are shown below.

$$\begin{aligned} {}^6\pi_g \quad \varphi(r) &= 858 \exp(-2.48r) - 675.8 \exp(-3.468r) \\ &\approx 678 \exp(-2.42r) \end{aligned} \quad (A5)$$

$$\begin{aligned} {}^6\pi_\mu \quad \varphi(r) &= 858 \exp(-2.48r) - 13.51.6 \exp(-3.468r) \\ &\approx 497 \exp(-2.32r) \end{aligned} \quad (A6)$$

$$\begin{aligned} {}^6\Sigma_g^+ \quad \varphi(r) &= 369 \exp(-2.46r) + 261.4 \exp(-2.977r) \\ &\approx 577 \exp(-2.57r) \end{aligned} \quad (A7)$$

$$\begin{aligned} {}^6\Sigma_\mu^+ \quad \varphi(r) &= 369 \exp(-2.46r) + 522 \exp(-2.977r) \\ &\approx 730 \exp(-2.60r) \end{aligned} \quad (A8)$$

The collision integrals for the exponential were obtained from the tables of [22], and are shown in Table V.

Multiplicity Two Potentials

The multiplicity two potentials include the ground state potential and the other low lying potential curves. The best fit to these potentials by a standard form is that of the Morse Potential.

$$\varphi = \epsilon \left\{ \exp \left(-2 \beta (r - r_e) \right) - 2 \exp \left(-\beta [r - r_e] \right) \right\} \quad (A9)$$

where

ϵ = the depth of the potential well

r_e = the position of the potential minimum

β = a parameter determining the shape of the potential

If the vibrational frequency of the ground state is known the parameter can be determined, see [24].

$$\beta = \sqrt{\frac{2\pi^2 c\mu}{\epsilon h}} \quad (A10)$$

From the knowledge of the vibrational frequency of the ${}^2\pi_\mu$, ${}^2\Sigma_g^+$, and ${}^2\Sigma_\mu^+$ potentials we obtain,

$${}^2\pi_\mu \quad \varphi(r) = 7.61 \left\{ \exp [-2 \times 2.48 (r - 1.1825)] \right. \\ \left. - 2 \exp [-2.48 (r - 1.1825)] \right\} \quad (A11)$$

$${}^2\Sigma_g^+ \quad \varphi(r) = 8.73 \left\{ \exp [-2 \times 2.68 (r - 1.1167)] \right. \\ \left. - 2 \exp [-2.68 (r - 1.1167)] \right\} \quad (A12)$$

$${}^2\Sigma_\mu^+ \quad \varphi(r) = 5.56 \left\{ \exp [-2 \times 3.678 (r - 1.0744)] \right. \\ \left. - 2 \exp [-3.678 (r - 1.0744)] \right\} \quad (A13)$$

It is found that the Morse curves for the ${}^2\Sigma_g^+$ and ${}^2\pi_\mu$ potentials give excellent fits to the curves of [25], but that the ${}^2\Sigma_\mu^+$ potential has an anomolous shape and is poorly fitted. Near the potential minimum the fit is good, but beyond 1.4 Å the potential is considerably deeper than the formula would indicate. The parameter β for the ${}^2\pi_g$ potential was determined by the relation

$$\beta = \frac{\ln(2)}{\sigma - r_e} \quad (A14)$$

where σ is the distance where the potential is zero. Hence,

$$\phi(r) = 2.35 \begin{cases} \exp[-2 \times 2.48 (r-1.48)] \\ -2 \exp[-2.48 (r-1.48)] \end{cases} \quad (A15)$$

Tables of collision integrals for Morse functions exist. The work of Smith and Munn, [26], was used, but there is also the older work of Lovell and Hirschfelder, [27]. The two are in some disagreement at lower energies. It was felt that in the later work Smith and Munn more care was taken to overcome mathematical inaccuracies. The tables given are very coarse and extensive interpolation in both temperature and the equivalent of the parameter β was necessary. Alternate calculations using Buckingham (6 exp) potential were made.

$$\begin{aligned} \phi(r) &= \begin{cases} \frac{\epsilon}{1 - \frac{6}{\alpha}} \left[\frac{6}{\alpha} \exp \left(\alpha \left[1 - \frac{r}{r_e} \right] \right) - \left(\frac{r_e}{r} \right)^6 \right] & r > r_{\max} \\ \infty & r < r_{\max} \end{cases} \\ & \quad (A16) \end{aligned}$$

Using α equal to twelve, significant differences of up to 25 percent, in the individual cross sections were found. Use of the Lennard Jones Potential would give even greater differences.

There was no really good way to handle the anomalously shaped $^2\Sigma^+_{\mu}$ potential. A Morse curve more closely fitting the far portion of the curve was used at the low energies and the results blended into the Morse potential which fits the near portion at the higher energies.

Multiplicity Four Potentials

The multiplicity four potentials consist of two very shallow regular potentials, the $^4\Sigma^+_g$ and $^4\Pi_{\mu}$, and two potentials having a small far maximum and a near minimum, the $^4\Sigma^+_{\mu}$ and $^4\Pi_g$.

Consider the $^4\Sigma_g^+$ potential. From [25] we have: $\epsilon = 0.2$ E. V., $r_e = 2.12$ A, and $\sigma = 1.89$ A. From eq. A14 we obtain $\beta = 3.0$. The far potential of [4] is,

$$^4\Sigma_g^+ \quad \phi(r) = -130.7 \exp(-2.977r) \quad (A20)$$

Taking the far limit of the Morse potential we see that the agreement for the coefficient in the exponential is good, but that there is almost a factor of two disagreement on the other coefficient. Further the results of [4] would deny the existence of a potential minimum beyond 1.95 A.

For the $^4\Pi_\mu$ potential we obtain the Morse potential,

$$^4\Pi_\mu \quad \phi(r) = .55 \left\{ \begin{array}{l} \exp[-2 \times 2.3(r-1.68)] \\ -2 \exp[-2.3 \times (r-1.68)] \end{array} \right\} \quad (A21)$$

This is not in good agreement with the results of [4]

$$^4\Pi_\mu \quad \phi(r) = -337.9 \exp(-3.468r) \quad r > 1.95A \quad (A22)$$

In both of the above cases final calculations were carried out using the Morse curve.

As previously mentioned the $^4\Pi_g$ potential has a far maximum and near minimum. These are .23 E. V. at 2.05 A and 1.0 E. V. at 1.54 A respectively according to Gilmore, [25]. The only potential investigated which has this type of behavior is the 12-6-3 potential, [28].

$$\phi(r) = 4\epsilon_o \left[\left(\frac{\sigma_o}{r} \right)^{12} - \left(\frac{\sigma_o}{r} \right)^6 + 8 \left(\frac{\sigma_o}{r} \right)^3 \right] \quad (A23)$$

Note that the potential is not "normalized" so that ϵ_o does not equal the depth of the potential minimum and σ_o is not the distance where the potential is zero. For $\delta = 0.25$ the 12-6-3 potential very well approximates

the ${}^4\pi_g$ potential. The tables do not extend sufficiently far down in energy. For temperatures corresponding to energies of less than 1/4 E.V. use is made of the exponential from of the potential of [4].

$${}^4\pi_g \quad \phi(r) = 337.9 \exp(-3.46r) \quad r > 1.95A \quad (A24)$$

The ${}^4\Sigma^+_u$ potential has the same maximum but a deeper minimum of 1.4 E.V. at 1.18A [25]. Unfortunately there is no 12-6-3 potential of similar proportions for which there are tables of collision integrals. The best that can be done is to take a Morse curve to fit the minimum and use it at higher temperatures, fit the low temperatures to the exponential of [4], and somehow interpolate at intermediate temperatures.

$${}^4\Sigma^+_u \quad \phi(r) = 1.4 \exp[-2 \times 5.2 (r - 1.18)] \quad r < 1.7 \quad (A25)$$

$$-2 \exp[-5.2 (r - 1.18)]$$

$$= 130.7 \exp(-2.977r) \quad r > 1.95 \quad (A26)$$

CHARGE INDUCED DIPOLE

At large separations the potential between the the nitrogen atom and ion will be the inverse fourth power charge induced dipole potential.

$$\phi = - \frac{\alpha e^2}{2r^4} \quad (A27)$$

Hence at the low temperatures the collision integral will be that due just to this longest range force.

$$\overline{\Omega(2,2)} = \frac{393}{T^{1/2}} \quad (\text{A28})$$

The difficulty is in obtaining the transition between the high temperature results just calculated and in the low temperature asymptote. This can not be done rigorously. From the following discussion estimates of the effect will be drawn.

References exist dealing with the effect of an inverse fourth power potential combined with repulsive and other attractive potentials [18] & [20]. Mason and Schamp [18] considered the 12-6-4 potential.

$$\phi = \frac{\epsilon}{2} \left[(1+\gamma) \left(\frac{re}{\gamma} \right)^{12} - 4\gamma \left(\frac{re}{\gamma} \right)^6 - 3(1-\gamma) \left(\frac{re}{\gamma} \right)^4 \right] \quad (\text{A29})$$

The parameter γ indicates the relative importance of the inverse sixth and fourth power potentials. If γ is zero, there is no inverse sixth power attraction and we have a 12-4 potential; if γ is one, there is no inverse fourth power attraction and we have the standard Lennard-Jones 12-6 potential. The 8-4 potential [20] gives qualitatively the same behavior as the 12-4 potential. If the temperature is nondimensionalized by the use of

$$T^* = \frac{kT}{\epsilon} \quad (\text{A30})$$

it is found that the collision integrals differs negligibly from the high temperature asymptote for $T^* > 4$, and differs negligibly from the low temperature asymptote, the charge induced dipole limit for $T^* < .04$.

The behavior in the intermediate region is determined by the nature of the other potentials. For the charge induced dipole and a pure repulsive potential, as one would have for the multiplicity six potentials, the collision integral in the intermediate region falls below the charge induced dipole and smoothly joins the high temperature asymptote from above. This is in contrast to the behavior of the charge exchange and charge induced dipole where the two asymptotes were joined with a smooth curve having no points of inflection [4]. The addition of other attractive potentials raises the curve in the intermediate region so that it may in part or whole be above the charge induced dipole asymptote.

The limitation in the use of the above references is that one can not consider the charge induced dipole as being a separate term added onto other potentials, except perhaps at large separation. For the multiplicity six potentials one can attempt to determine the minimum produced by the charge induced dipole by adding it to the exponential repulsion and finding the location and magnitude of the minimum, see Table VII. Such procedure is possible. However this seems to underestimate the region of importance of the charge induced dipole. For the other potentials not even this crude procedure is applicable. The final corrections made to include the charge induced dipole are the result graphical interpolation of the high and low temperature limits guided by the limited knowledge available.

TABLE 1. ELECTRONIC LEVELS OF POSSIBLE IMPORTANCE

	Designation	Energy (E/k in °K)	Multiplicity
N	$2^3_2 \text{ } ^4\text{S}^0$	0	4
	$2^3_2 \text{ } ^2\text{D}^0$	27, 650	10
	$2^3_2 \text{ } ^2\text{P}^0$	41, 480	6
N ⁺	$2^2_2 \text{ } ^3\text{P}$	0	1
		70	3
		189	5
	$2^2_2 \text{ } ^1\text{D}$	21, 900	5
N ₂	$1^+_1 \Sigma_g$	0	1

TABLE II. THE INTERACTIONS

	N_2	N	N^*	N^+	e
N_2	neutral-neutral experimental results at lower temperatures	neutral-neutral	same as N - N_2 ?	charge induced dipole	experimental results
N		neutral-neutral 4 potential curves	excitation exchange	charge exchange charge induced dipole	Theoretical quantum mech. results
N^*			same as N - N ?	same as N - N^+ ?	same as e - N ?
N^+				Coulombic	Coulombic
e					Coulombic

TABLE III. POTENTIALS FOR N - N⁺ INTERACTION (4)

State	Statistical Weight (total of 72)	Type of Potential
${}^6\pi_g$	12	repulsive - known only for $r > 1.95 \text{ \AA}$
${}^6\pi_u$	12	repulsive - known only for $r > 1.95 \text{ \AA}$
${}^6\Sigma_g^+$	6	repulsive - known only for $r > 1.95 \text{ \AA}$
${}^6\Sigma_u^+$	6	repulsive - known only for $r > 1.95 \text{ \AA}$
${}^4\pi_g$	8	near minimum and far maximum
${}^4\Sigma_u^+$	4	near minimum and far maximum
${}^4\pi_u$	8	shallow potential well
${}^4\Sigma_g^+$	4	very shallow potential well
${}^2\pi_g$	4	attractive - repulsive
${}^2\pi_u$	4	strong attractive-repulsive
${}^2\Sigma_u^+$	2	strong attractive-repulsive anomalous shape
${}^2\Sigma_g^+$	2	strong attractive-repulsive

TABLE IV. LONG RANGE BEHAVIOR OF THE $N - N^+$ POTENTIALS
NEGLECTING THE CHARGE INDUCED DIPOLE [4]

Multiplicity	$\Sigma_{\mu, g}^+$	$\pi_{\mu, g}$
2	$\frac{2}{3} J\pi \pm \tilde{Q}$	$\frac{1}{3} (J'\pi + J'\sigma) \pm \tilde{Q}'$
4	$\pm \tilde{Q}$	$\pm \tilde{Q}'$
6	$-2 J\pi \pm \tilde{Q}$	$-(J'\pi + J'\sigma) \pm \tilde{Q}'$

TABLE V. COLLISION CROSS SECTIONS FOR THE $N - N^+$ POTENTIALS
 $\Omega^{(2,2)}$ NEGLECTING THE CHARGE INDUCED DIPOLE

	kT (in E. V.)								
	5.0	2.5	1.0	.50	.25	.10	.050	.025	.010
$6\pi_u$	4.03	5.37	7.41	9.17	11.11	13.95	16.31	18.85	22.48
$6\pi_g$	4.23	5.54	7.52	9.21	11.07	13.77	16.02	18.42	21.85
$6\Sigma_u^+$	3.79	4.93	6.62	8.15	9.78	12.14	14.10	16.20	19.20
$6\Sigma_g^+$	3.51	4.63	6.34	7.80	9.42	11.77	13.73	15.83	18.82
$4\pi_u$	1.26	1.62	2.28	4.36	6.00	8.75	10.50	12.40	15.20
$4\pi_g$	1.39	1.65	2.32	3.06	4.48	5.69	6.70	7.79	19.36
$4\Sigma_u^+$	1.04	1.32	1.97	2.60	4.58	6.02	7.24	8.58	10.50
$4\Sigma_g^+$	2.43	2.72	3.36	3.87	4.72	7.15	8.60	9.15	11.40
$2\pi_u$	2.62	4.31	6.50	8.65	9.60	11.45	13.00	14.60	17.90
$2\pi_g$	2.00	2.70	4.63	6.61	8.97	12.51	13.1	15.6	19.1
$2\Sigma_u^+$	1.69	2.9	4.9	6.3	7.6	9.7	11.3	13.4	16.7
$2\Sigma_g^+$	2.59	4.00	5.92	7.33	8.59	10.38	11.82	13.49	15.70

TABLE VI. WEIGHTED COLLISION CROSS SECTIONS FOR THE $N - N^+$
POTENTIALS $\Omega^{(2,2)}$ NEGLECTING THE CHARGE INDUCED DIPOLE

		kT (In E. V.)								
	Statistical Weight (in 72ths)	5.0	2.5	1.0	.50	.25	.10	.05	0.25	.010
${}^6\pi_u$	12	.671	.894	1.236	1.529	1.852	2.33	2.72	3.14	3.75
${}^6\pi_g$	12	.705	.923	1.253	1.535	1.844	2.30	2.67	3.07	3.64
${}^6\Sigma^+_u$	6	.316	.411	.556	.679	.815	1.01	1.18	1.35	1.60
${}^6\Sigma^+_g$	6	.292	.386	.528	.650	.785	.981	1.14	1.32	1.57
${}^4\pi_u$	8	.140	.180	.253	.484	.667	.972	1.17	1.30	1.69
${}^4\pi_g$	8	.154	.183	.258	.340	.498	.632	.744	.865	1.04
${}^4\Sigma^+_u$	4	.055	.073	.109	.144	.254	.334	.402	.477	.583
${}^4\Sigma^+_g$	4	.135	.151	.187	.215	.262	.397	.478	.508	.633
${}^2\pi_u$	4	.146	.239	.361	.458	.533	.636	.722	.811	.995
${}^2\pi_g$	4	.111	.150	.263	.367	.498	.695	.728	.867	1.06
${}^2\Sigma^+_u$	2	.047	.085	.136	.175	.211	.269	.314	.372	.464
${}^2\Sigma^+_g$	2	.072	.111	.164	.204	.239	.288	.328	.375	.436
total		2.84	3.79	5.30	6.78	8.46	10.84	12.6	14.5	23.5
repulsive contribution		1.98	2.61	3.57	4.39	5.30	6.62	7.71	8.88	10.6

TABLE VII. POTENTIAL MINIMUM FOR THE REPULSIVE POTENTIALS
WITH THE CHARGE INDUCED DIPOLE

	r_e	ϵ (E. V.)	$\frac{\epsilon}{k}$ ($^{\circ}$ K)
${}^6\pi_g$	4.88	.00955	110
${}^6\pi_u$	4.88	.00958	112
${}^6\Sigma_g^+$	4.40	.0137	159
${}^6\Sigma_u^+$	4.40	.0132	153

TABLE VIII. COLLISION CROSS SECTIONS FOR THE $N - N^+$ POTENTIALS
 $\Omega(2,2)$ INCLUDING THE CHARGE INDUCED DIPOLE

	kT (In E.V.)								
	5.0	2.5	1.0	.50	.25	.10	.050	.025	.010
${}^6\pi_u$								26.0	36.6
${}^6\pi_g$	no change							26.0	36.6
${}^6\Sigma^+_u$							14.3	25.0	36.6
${}^6\Sigma^+_g$							14.3	25.0	36.6
${}^4\pi_u$						8.9	13.0	20.0	36.6
${}^4\pi_g$	no change					6.6	9.0	17.5	36.6
${}^4\Sigma^+_u$						6.6	9.0	17.5	36.6
${}^4\Sigma^+_g$							9.6	17.5	36.6
${}^2\pi_u$						12.2	17.0	22.2	36.6
${}^2\pi_g$						12.2	17.0	22.2	36.6
${}^2\Sigma^+_u$						12.2	17.0	22.2	36.6
${}^2\Sigma^+_g$						12.2	17.0	22.2	36.6

TABLE IX. WEIGHTED COLLISION CROSS SECTIONS FOR THE $N - N^+$
POTENTIALS $\Omega^{(2,2)}$ INCLUDING CHARGE INDUCED DIPOLE

		kT (In E. V.)								
Statistical Weight		5.0	.25	.10	.50	.25	.10	.05	.025	.010
$6\pi_u$	12	.671	.894	1.236	1.529	1.852	2.33	2.72	4.33	6.10
$6\pi_g$	12	.705	.923	1.253	1.535	1.844	2.30	2.72	4.33	6.10
$6\Sigma^+_u$	6	.316	.411	.556	.679	.815	1.01	1.19	2.08	3.05
$6\Sigma^+_g$	6	.292	.386	.528	.650	.785	.98	1.19	2.08	3.05
$4\pi_u$	8	.140	.180	.253	.484	.667	.99	1.44	2.22	9.06
$4\pi_g$	8	.154	.183	.258	.340	.498	.73	1.00	1.94	4.06
$4\Sigma^+_u$	4	.055	.073	.109	.144	.254	.37	.50	.97	2.02
$4\Sigma^+_g$	4	.135	.151	.187	.215	.262	.10	.53	.97	2.02
$2\pi_u$	4	.146	.239	.361	.458	.533	.68	.44	1.23	2.02
$2\pi_g$	4	.111	.150	.263	.367	.498	.68	.44	1.23	2.02
$2\Sigma^+_u$	2	.047	.085	.136	.175	.211	.34	.47	.62	1.01
$2\Sigma^+_g$	2	.072	.111	.164	.204	.239	.34	.47	.62	1.01
total		2.84	3.79	5.30	6.78	8.46	11.2	14.1	27.6	36.6
repulsive		1.98	2.61	3.57	4.39	5.30	6.62	7.8	12.8	18.3

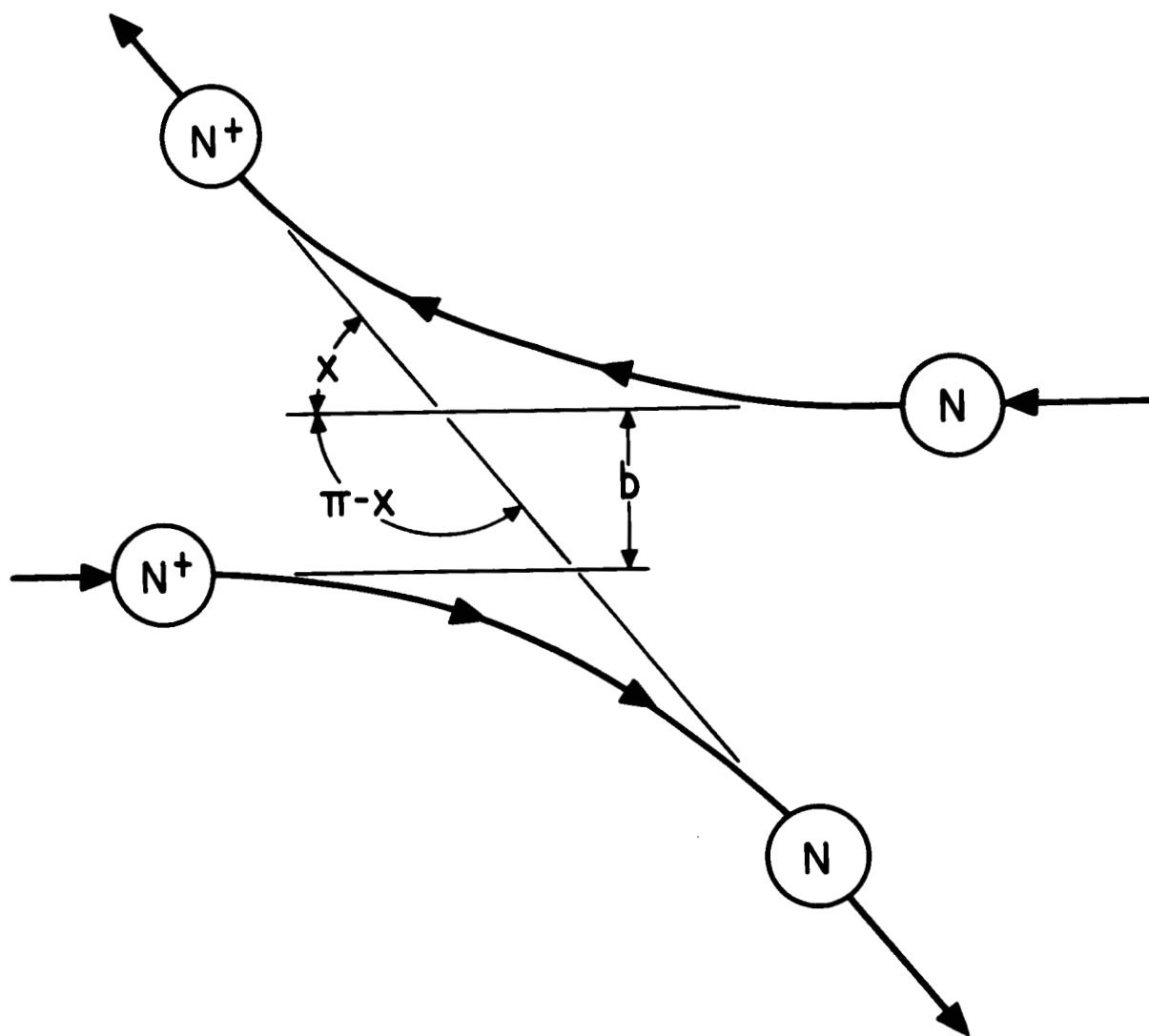


Figure 1. A Symmetric Charge Exchange Collision

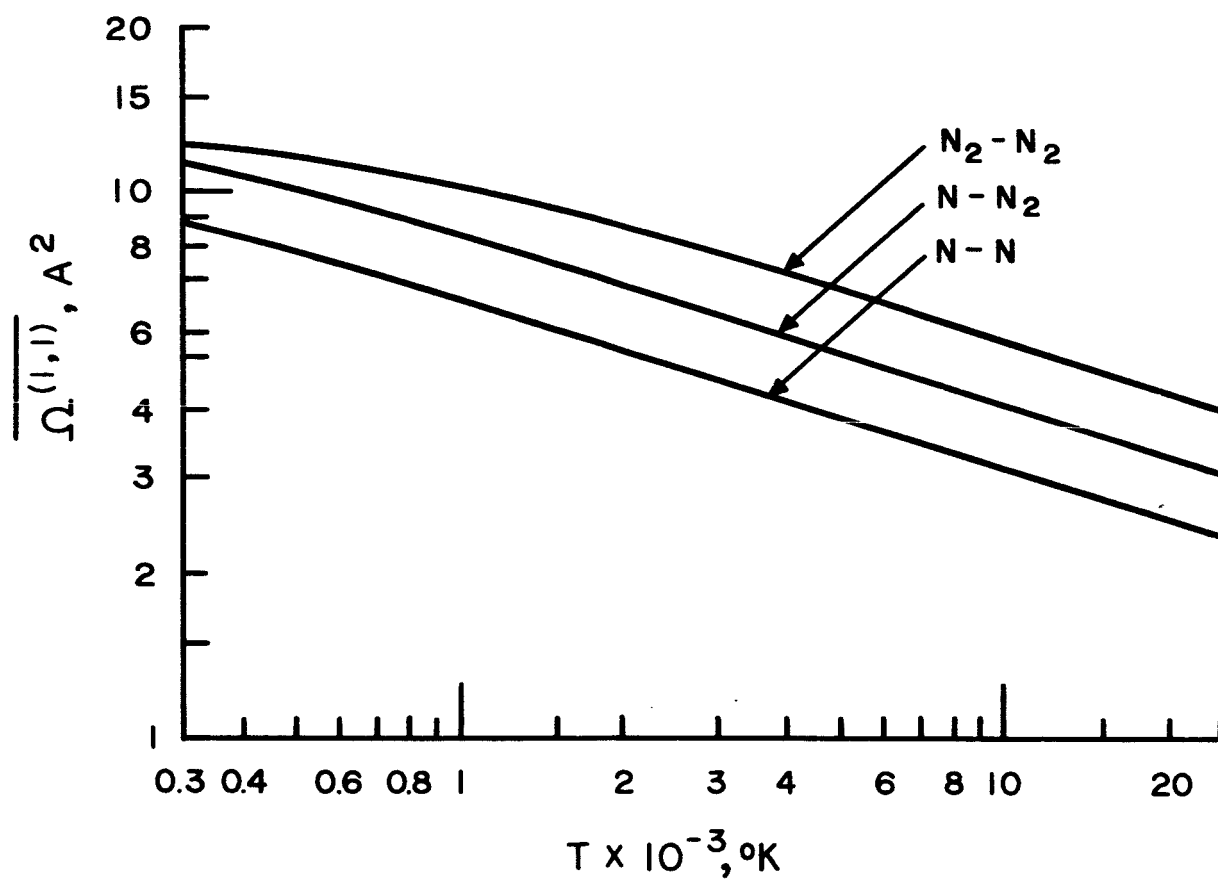


Figure 2. Neutral-Neutral $\overline{\Omega^{(1,1)}}$ Collision Integrals

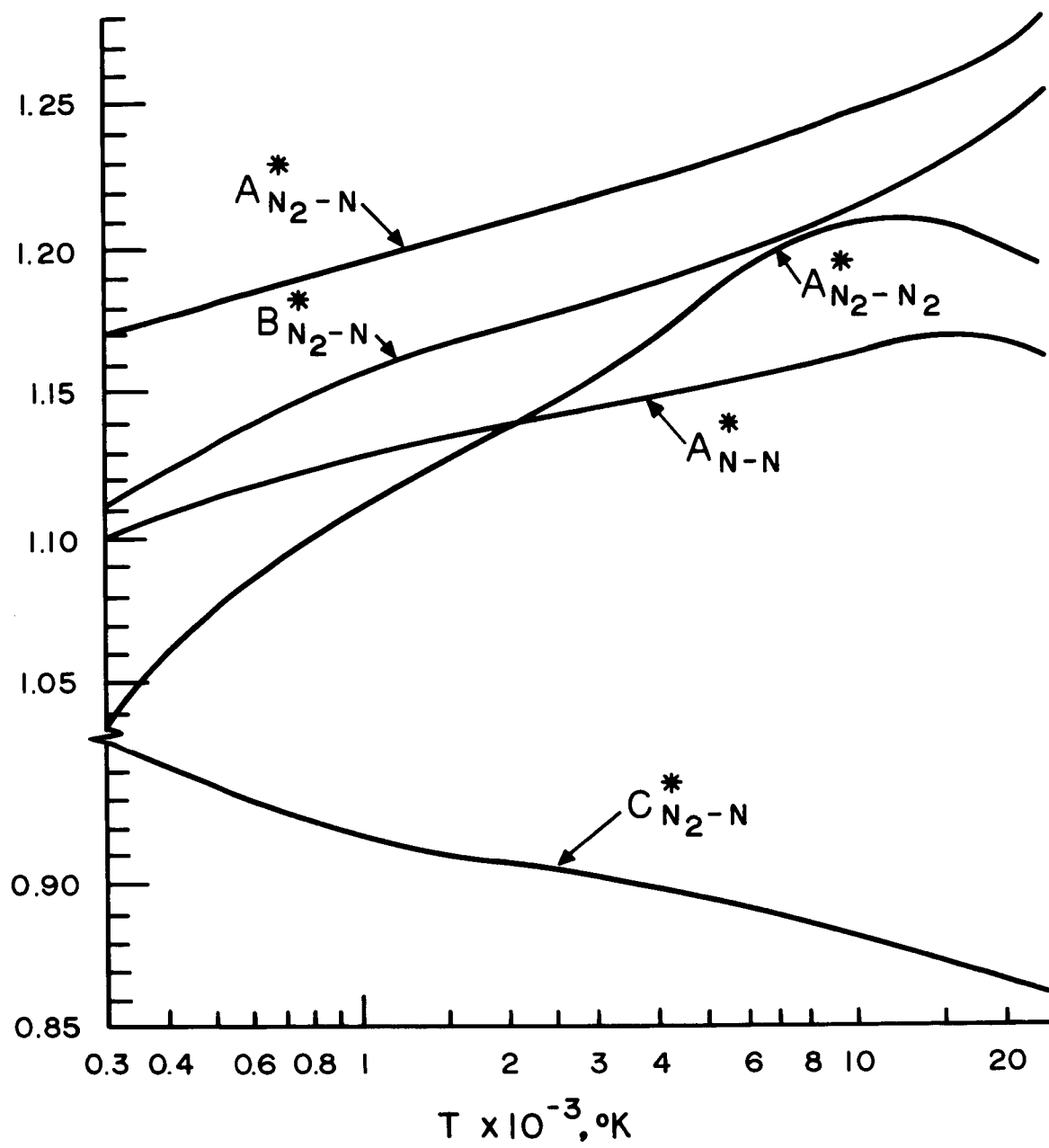


Figure 3. Neutral-Neutral A^* , B^* , C^*

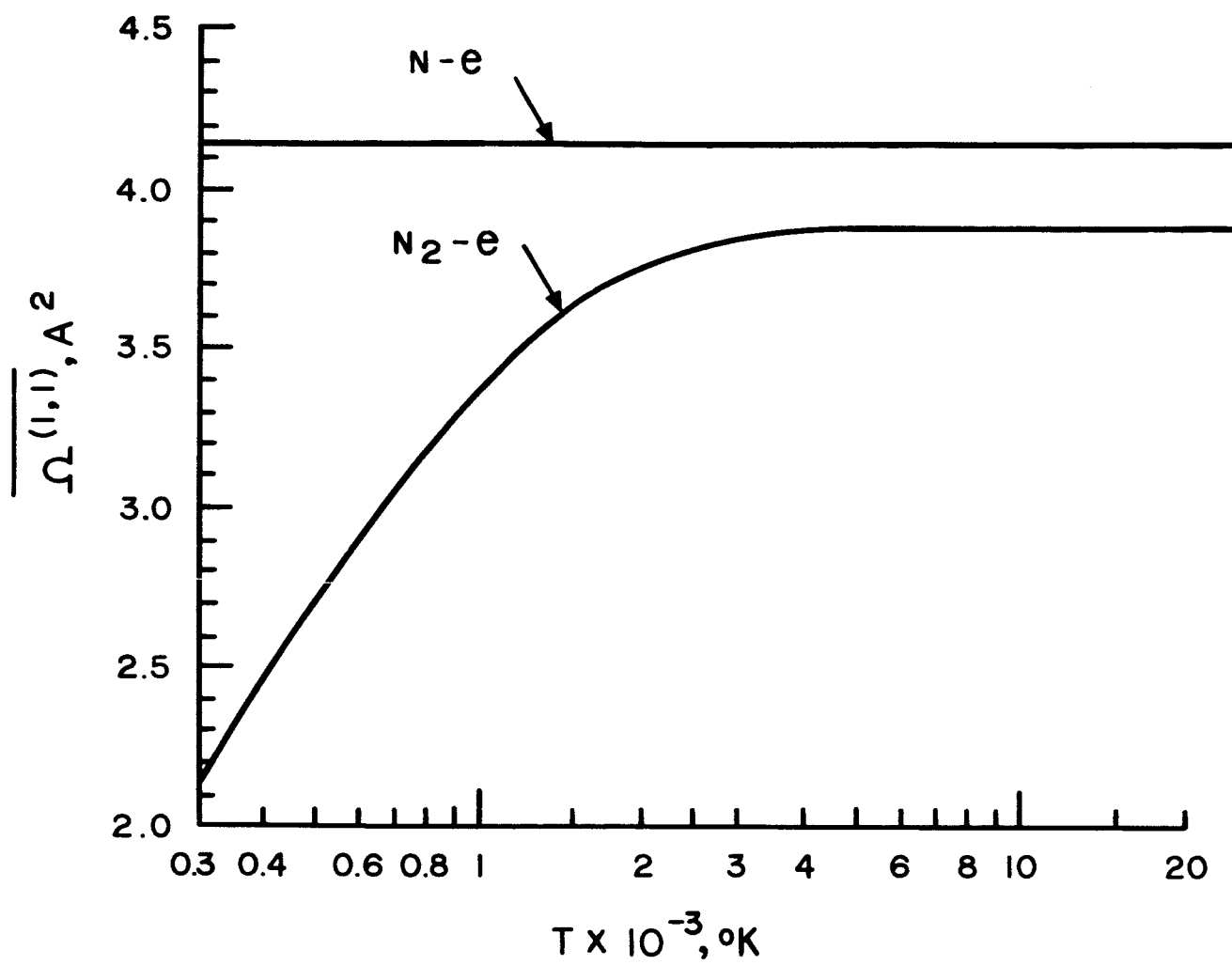


Figure 4. The e-N₂, e-N $\overline{\Omega^{(1,1)}}$ Collision Integral

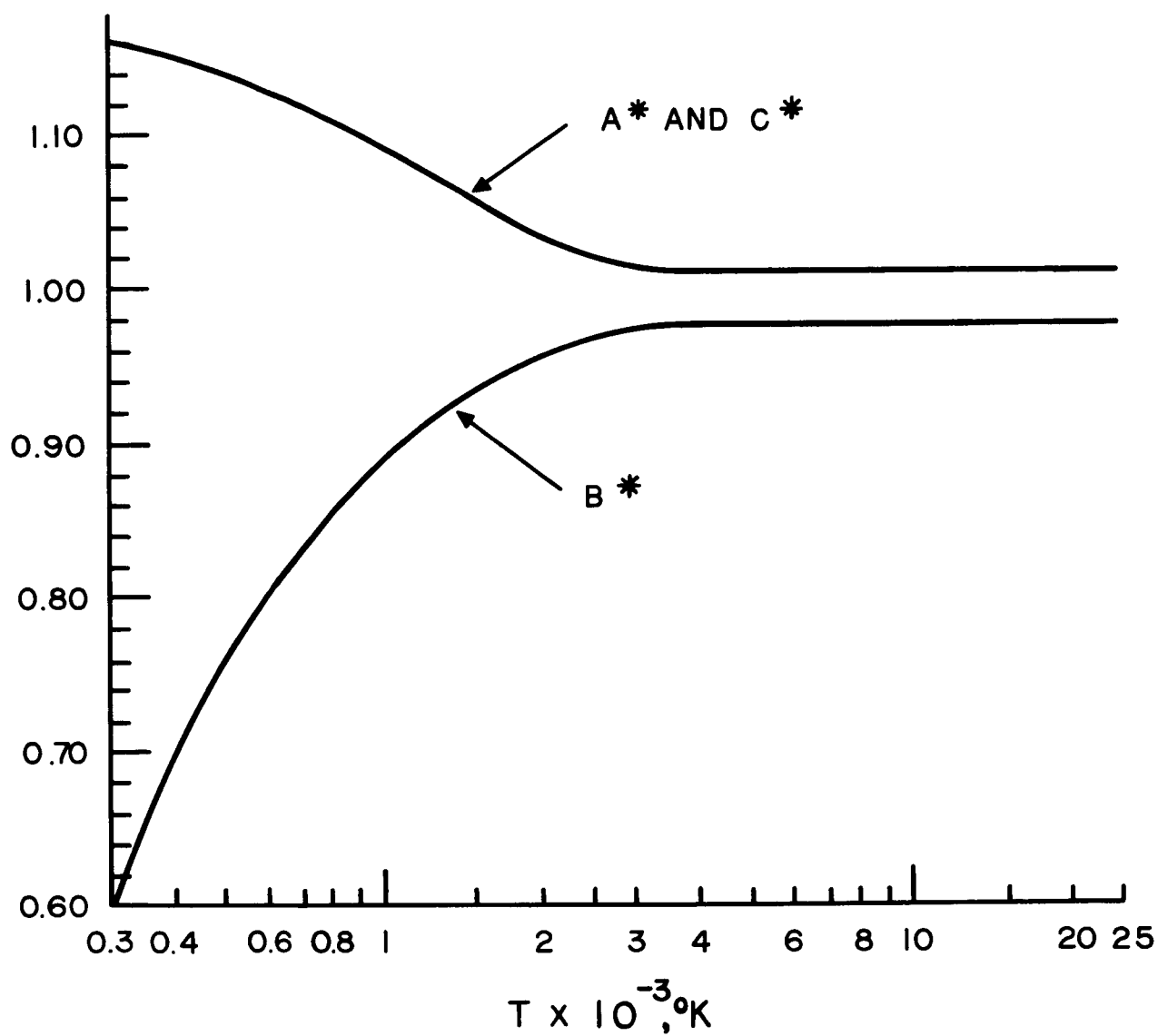


Figure 5. The e-N₂, A*, B*, C*

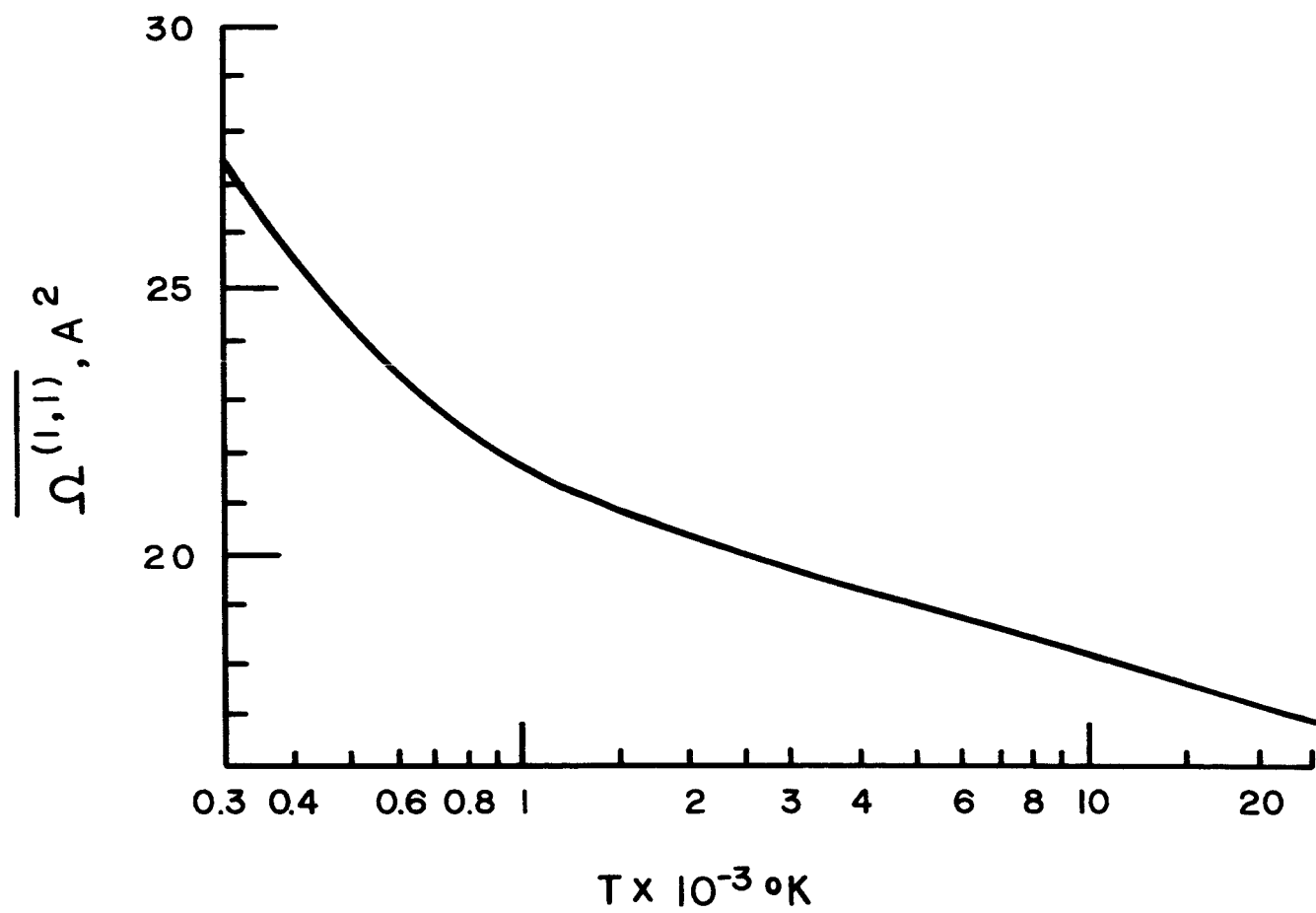


Figure 6. The $N-N^+ \overline{\Omega^{(1,1)}}$ Collision Integral (1)

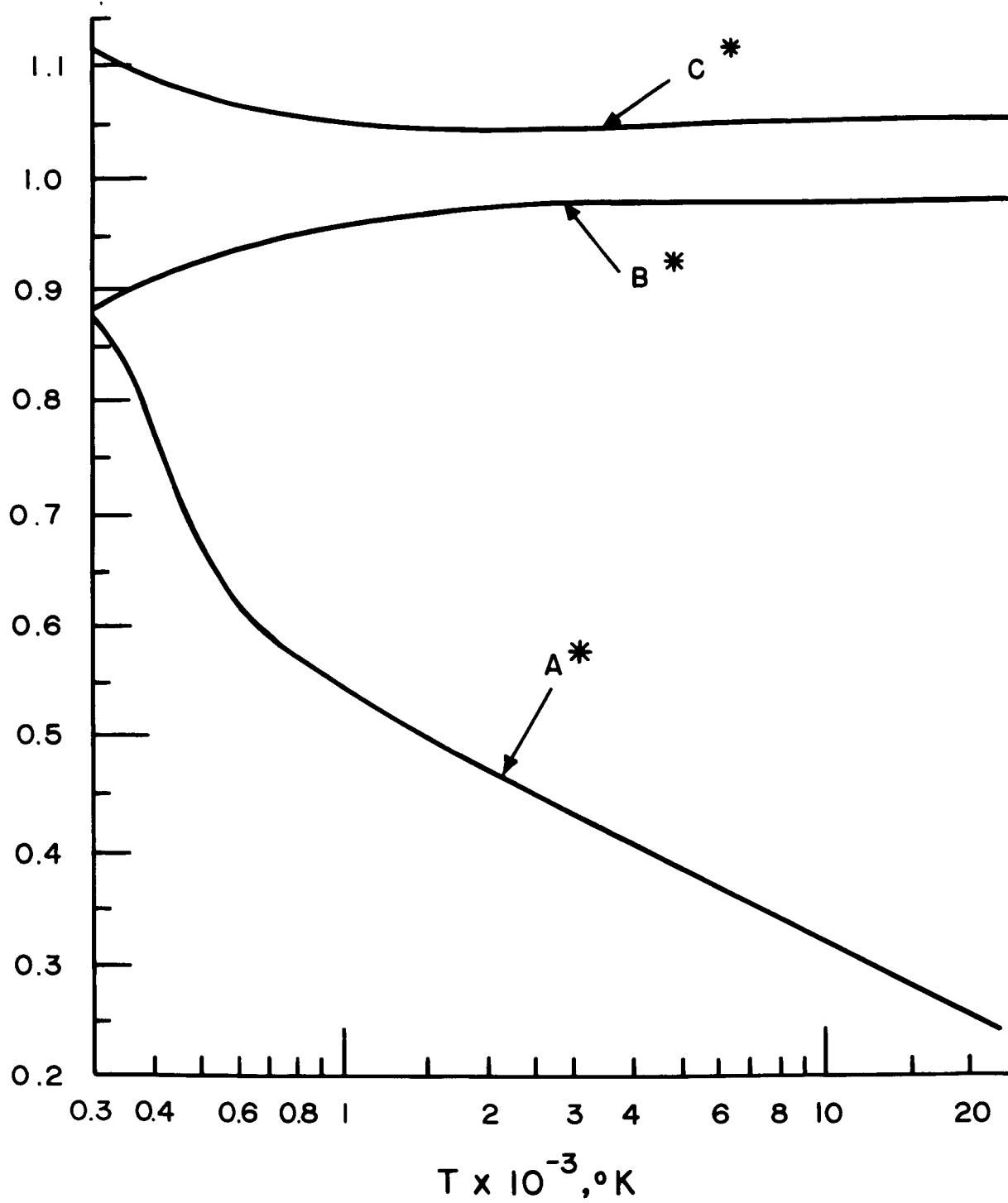


Figure 7. The $N-N^+$ A*, B*, C*

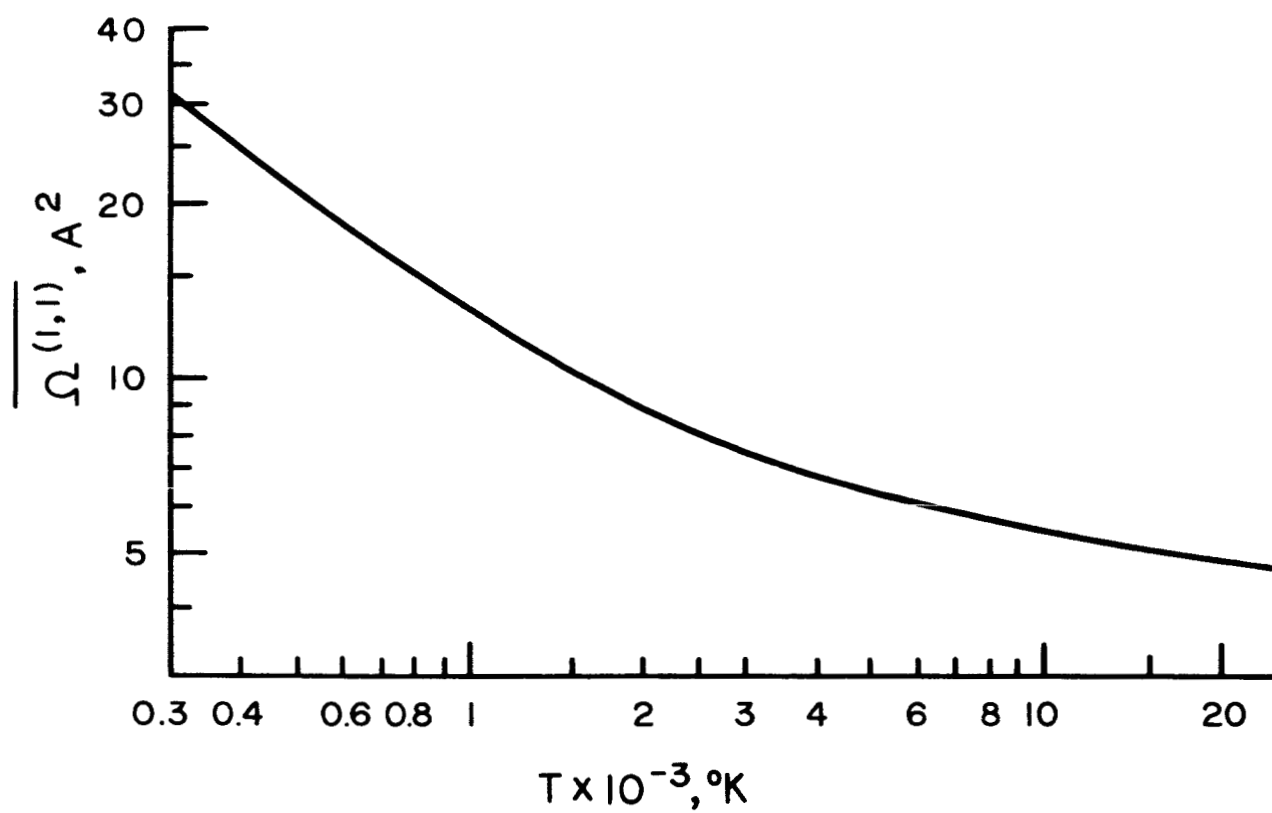


Figure 8. The $N^+ - N_2$ $\overline{\Omega^{(1,1)}}$ Collision Integral

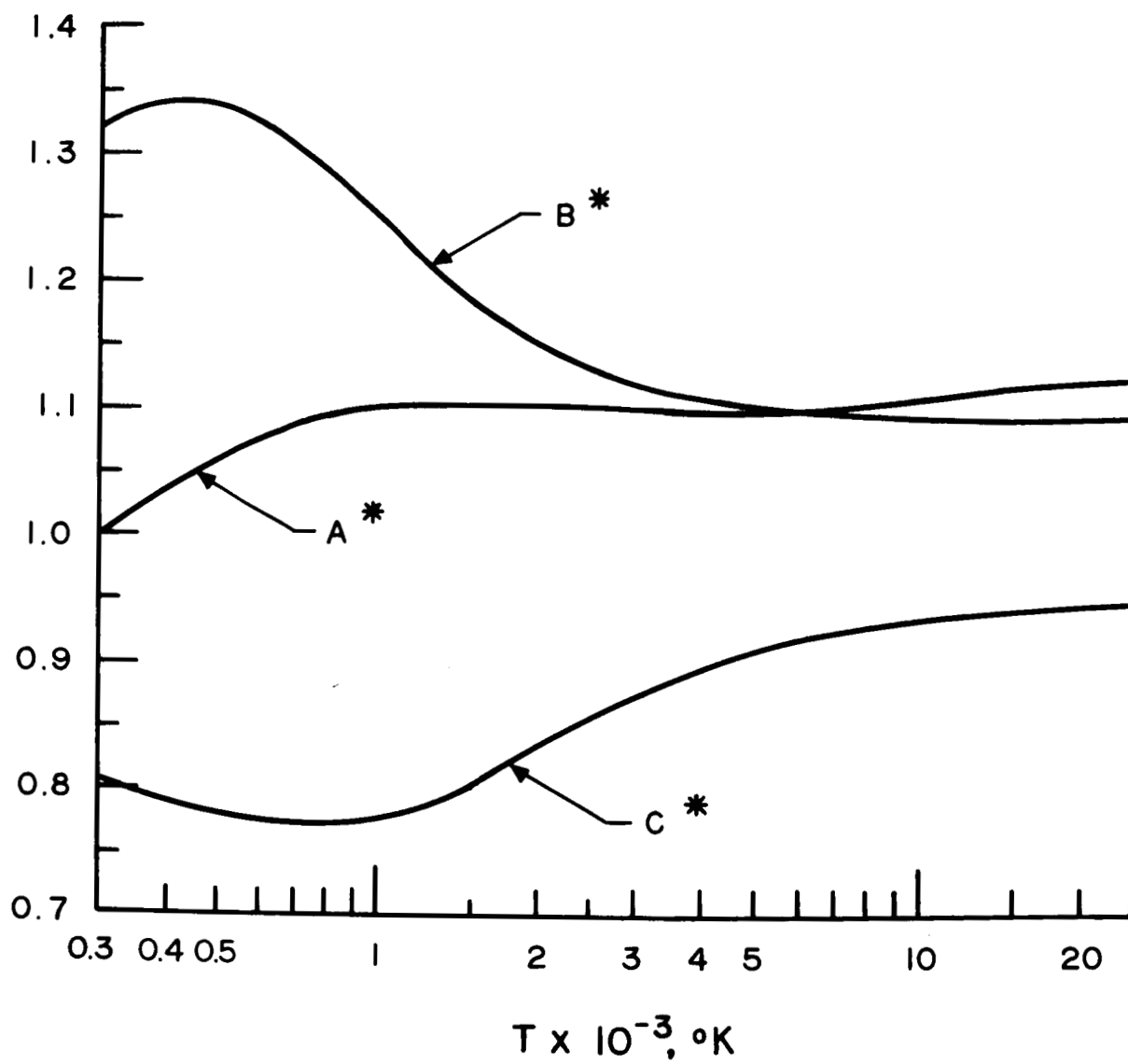


Figure 9. The $\text{N}^+ - \text{N}_2$ A*, B*, C*

TECHNICAL INFORMATION SERIES

AUTHOR M. P. Sherman	SUBJECT CLASSIFICATION Transport Properties	NO. R65 SD 43
		DATE July, 1965
TITLE Transport Properties of Partially Ionized Nitrogen I. The Collision Integrals		G. E. CLASS I
		GOV. CLASS U
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<p>SUMMARY</p> <p>This report is the first of two concerned with the computation of the transport properties of nitrogen up to temperatures where there is single ionization. In this report the collision integrals are obtained using the best available information. In particular, use is made of the recent work of Knof, Mason, and Vanderslice for the charge exchange cross section of the $N - N^+$ interaction. Calculations were performed to obtain the $\Omega^{(2,2)}$ collision integral for the $N - N^+$ interaction.</p> <p>In a second report, transport properties will be computed by a new method which permits rapid calculation of the transport properties of multicomponent mixtures valid up to the second approximation of the variational scheme of Hirschfelder, et. al.</p>		
<p>KEY WORDS</p> <p>Single ionization, Transport properties, Collisional integrals</p>		

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